

SCULPTURED THIN FILMS: ACCOMPLISHMENTS AND EMERGING USES

Akhlesh Lakhtakia¹

CATMAS — Computational and Theoretical Materials Sciences Group,
Department of Engineering Science and Mechanics,
Pennsylvania State University, University Park, PA 16802–6812, USA

Abstract

Sculptured thin films (STFs) are nano-engineered materials whose columnar morphology is tailored to elicit desired optical responses upon excitation. Two canonical forms of STFs have been identified. Linear constitutive relations for general STFs as unidirectionally nonhomogeneous (continuously or piecewise uniformly) and locally bianisotropic continua are presented, along with a 4×4 matrix ordinary differential equation for wave propagation therein. A nominal model for the macroscopic properties of linear STFs is devised from nanoscopic considerations. The accomplished implementation of STFs as circular polarization filters and spectral hole filters is discussed, as also are emerging applications such as bioluminescence sensors and optical interconnects.

Keywords: Bianisotropic materials, Biochips, Chiral optics, Local homogenization, Optical devices, Sculptured thin films, Sensors

1. Introduction

Shortly after the conceptualization of helicoidal bianisotropic mediums (HBMs) in 1993 by Lakhtakia and Weiglhofer [1], the basis of realizing these materials using thin-film technology was enunciated by Lakhtakia and Messier [2]. Verification by Robbie *et al.* [3] soon followed, although a few years later I came across a pioneering and essentially comprehensive but obscure precedent reported by Young and Kowal in 1959 [4]. The general concept of sculptured thin films (STFs) emerged naturally, and was presented in August 1995 by Lakhtakia and Messier to a group of thin-film researchers assembled at Penn State [5]. The topic has enjoyed considerable growth in the ensuing years, chiefly in theory initially, but now in experiments and applications as well [6].

The nanostructure of STFs comprises clusters of 3–5 nm diameter and arranged to form parallel columns that are bent in some fanciful forms with feature size 30 nm or larger. Accordingly, a STF is a unidirectionally nonhomogeneous continuum with direction-dependent properties at visible and infrared wavelengths. A multi-section STF can thus be conceived of as an optical circuit that can be integrated with electronic circuitry on a chip. Being porous, a STF can act as a sensor of fluids and can be impregnated with liquid crystals for switching applications

¹Tel. +1 814 863 4319; Fax. +1 814 863 7967; E-mail: AXL4@psu.edu

too. Application as low-permittivity barrier layers in electronic chips as well as for solar cells has also been suggested. During the last five year, several physical vapor deposition techniques have emerged for manufacturing STFs, and the first optical applications saw the light of the day in 1999.

The following is a brief review of (i) the electromagnetic field equations, (ii) a nominal nano-scopic-to-continuum model, (iii) realized optical applications, and (iv) emerging applications of STFs. A large part of the work reviewed here is due to my collaborators, my students and me. For the essentials of the fabrication techniques, the interested reader is enjoined to read the gem of a paper that Young & Kowal wrote [4]. For details of the modern versions of the Young-Kowal technique, reference is made to Lakhtakia & Messier [7], Hodgkinson & Wu [8], Messier *et al.* [9], and Malac & Egerton [10]. The materials that can be deposited as STFs range from insulators to semiconductors to metals, thereby indicating the versatility of STF technology.

2. Electromagnetic field equations

2.1 Linear constitutive equations

Let the z axis of a cartesian coordinate system be aligned parallel to the direction of nonhomogeneity. By definition, the morphology of a simple STF in any plane $z = z_1$ can be made to coincide with the morphology in another plane $z = z_2$ with the help of a suitable rotation. In other words, the *local* morphology is spatially uniform, but the *global* morphology is unidirectionally nonhomogeneous. Naturally, this leads to the concept of local or *reference constitutive properties* of the STF. The global constitutive properties of the STF can be connected to the local ones by means of rotation operators.

The frequency-domain constitutive relations of a chosen STF are therefore defined as follows:

$$\mathbf{D}(\mathbf{r}, \omega) = \epsilon_0 \underline{\underline{S}}(z) \cdot \left[\underline{\underline{\epsilon}}_{ref}(\omega) \cdot \underline{\underline{S}}^T(z) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\alpha}}_{ref}(\omega) \cdot \underline{\underline{S}}^T(z) \cdot \mathbf{H}(\mathbf{r}, \omega) \right], \quad (1)$$

$$\mathbf{B}(\mathbf{r}, \omega) = \mu_0 \underline{\underline{S}}(z) \cdot \left[\underline{\underline{\beta}}_{ref}(\omega) \cdot \underline{\underline{S}}^T(z) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\mu}}_{ref}(\omega) \cdot \underline{\underline{S}}^T(z) \cdot \mathbf{H}(\mathbf{r}, \omega) \right]. \quad (2)$$

In these relations, $\epsilon_0 = 8.854 \times 10^{-12}$ F/m and $\mu_0 = 4\pi \times 10^{-7}$ H/m are the permittivity and the permeability of vacuum, respectively, while the superscript T denotes the transpose. Whereas the 3×3 dyadics $\underline{\underline{\epsilon}}_{ref}(\omega)$ and $\underline{\underline{\mu}}_{ref}(\omega)$ represent the *reference* dielectric and magnetic properties, respectively, the 3×3 dyadics $\underline{\underline{\alpha}}_{ref}(\omega)$ and $\underline{\underline{\beta}}_{ref}(\omega)$ delineate the *reference* magneto-electric properties. The angular frequency is denoted by ω , and an $\exp(-i\omega t)$ time-dependence is implicit.

Any STF for optical and/or infrared applications comprises identical columns of 20–100 nm diameter. Nominally, all columns twist and bend identically as z changes, which feature is captured by the rotation dyadic $\underline{\underline{S}}(z)$. This 3×3 dyadic is some composition of the following

three elementary rotation 3×3 dyadics:

$$\underline{\underline{S}}_x(z) = \mathbf{u}_x \mathbf{u}_x + (\mathbf{u}_y \mathbf{u}_y + \mathbf{u}_z \mathbf{u}_z) \cos \xi(z) + (\mathbf{u}_z \mathbf{u}_y - \mathbf{u}_y \mathbf{u}_z) \sin \xi(z), \quad (3)$$

$$\underline{\underline{S}}_y(z) = \mathbf{u}_y \mathbf{u}_y + (\mathbf{u}_x \mathbf{u}_x + \mathbf{u}_z \mathbf{u}_z) \cos \tau(z) + (\mathbf{u}_z \mathbf{u}_x - \mathbf{u}_x \mathbf{u}_z) \sin \tau(z), \quad (4)$$

$$\underline{\underline{S}}_z(z) = \mathbf{u}_z \mathbf{u}_z + (\mathbf{u}_x \mathbf{u}_x + \mathbf{u}_y \mathbf{u}_y) \cos \zeta(z) + (\mathbf{u}_y \mathbf{u}_x - \mathbf{u}_x \mathbf{u}_y) \sin \zeta(z). \quad (5)$$

The angular functions of z in these equations may be specified piecewise, if necessary; while \mathbf{u}_x , \mathbf{u}_y and \mathbf{u}_z are the three cartesian unit vectors.

All columns in a STF are parallel to each other in any xy plane, which characteristic is incorporated in $\underline{\underline{\epsilon}}_{ref}(\omega)$, $\underline{\underline{\mu}}_{ref}(\omega)$, $\underline{\underline{\alpha}}_{ref}(\omega)$ and $\underline{\underline{\beta}}_{ref}(\omega)$. Suppose $\underline{\underline{S}}(0) = \underline{\underline{I}}$, the 3×3 identity dyadic, so that the plane $z = 0$ is the reference plane. Let \mathbf{u}_τ denote a unit vector that is tangential at $z = 0$ to any column. Two other unit vectors are defined from the shape of that column as follows: the unit normal vector \mathbf{u}_n such that $\mathbf{u}_n \cdot \mathbf{u}_\tau = 0$, and the unit binormal vector $\mathbf{u}_b = \mathbf{u}_\tau \times \mathbf{u}_n$. Both \mathbf{u}_τ and \mathbf{u}_n are conveniently chosen to lie in the plane $y = 0$; hence, \mathbf{u}_b is either parallel or anti-parallel to \mathbf{u}_y . With an angle of rise χ specified, we set

$$\mathbf{u}_\tau = \mathbf{u}_x \cos \chi + \mathbf{u}_z \sin \chi, \quad \mathbf{u}_n = -\mathbf{u}_x \sin \chi + \mathbf{u}_z \cos \chi, \quad \mathbf{u}_b = -\mathbf{u}_y, \quad (6)$$

these three unit vectors forming a right-handed coordinate system. Typically, the angle of rise $\chi \in (0^\circ, 90^\circ]$.

Extensive experimental research on the uniform columnar thin films [11] shows that the prescription

$$\underline{\underline{\epsilon}}_{ref}(\omega) = \epsilon_a(\omega) \mathbf{u}_n \mathbf{u}_n + \epsilon_b(\omega) \mathbf{u}_\tau \mathbf{u}_\tau + \epsilon_c(\omega) \mathbf{u}_b \mathbf{u}_b \quad (7)$$

is appropriate. This $\underline{\underline{\epsilon}}_{ref}(\omega)$ is a biaxial dyadic; and the simplification $\epsilon_c(\omega) = \epsilon_a(\omega)$ is appropriate for a locally uniaxial STF in the present context. If necessary, a gyrotropic term $\epsilon_g(\omega) \mathbf{u}_g \times \underline{\underline{I}}$ may be appended to the right side of (7), with \mathbf{u}_g as some unit vector. The forms of $\underline{\underline{\mu}}_{ref}(\omega)$, $\underline{\underline{\alpha}}_{ref}(\omega)$ and $\underline{\underline{\beta}}_{ref}(\omega)$ are similar to those of $\underline{\underline{\epsilon}}_{ref}(\omega)$.

Two canonical forms of STFs can be identified. For sculptured nematic thin films (SNTFs), either $\underline{\underline{S}}(z) = \underline{\underline{S}}_x(z)$ or $\underline{\underline{S}}(z) = \underline{\underline{S}}_y(z)$. The columnar morphology is essentially 2-dimensional, lying in either the xz plane or the yz plane. On the other hand, thin-film helicoidal bianisotropic mediums (TFHBMs) are endowed with 3-dimensional morphology, because $\underline{\underline{S}}(z) = \underline{\underline{S}}_z(z)$. Although TFHBMs need not be periodically nonhomogeneous along the z axis, it is easy to fabricate them with periods chosen anywhere between 200 nm and 2000 nm. Of course, combinations of the two canonical forms as well as cascades of multiple sections are possible, and add to the attraction of STFs.

2.2 Wave propagation

Electromagnetic wave propagation in a STF is best handled using 4×4 matrixes and column 4-vectors. At any given frequency, the following spatial Fourier representation of the electric

and the magnetic field phasors is useful:

$$\left. \begin{aligned} \mathbf{E}(\mathbf{r}, \omega) &= \mathbf{e}(z, \kappa, \psi_{inc}, \omega) \exp [i\kappa(x \cos \psi_{inc} + y \sin \psi_{inc})] \\ \mathbf{H}(\mathbf{r}, \omega) &= \mathbf{h}(z, \kappa, \psi_{inc}, \omega) \exp [i\kappa(x \cos \psi_{inc} + y \sin \psi_{inc})] \end{aligned} \right\}. \quad (8)$$

Substitution of the foregoing representation into the source-free Maxwell curl postulates, $\nabla \times \mathbf{E}(\mathbf{r}, \omega) = i\omega \mathbf{B}(\mathbf{r}, \omega)$ and $\nabla \times \mathbf{H}(\mathbf{r}, \omega) = -i\omega \mathbf{D}(\mathbf{r}, \omega)$, followed by the use of the constitutive relations (1) and (2) leads to four ordinary differential equations and two algebraic equations. The components $e_z(z, \kappa, \psi_{inc}, \omega)$ and $h_z(z, \kappa, \psi_{inc}, \omega)$ are then eliminated to obtain the 4×4 matrix differential equation [12]

$$\frac{d}{dz} [\mathbf{f}(z, \kappa, \psi_{inc}, \omega)] = i[\mathbf{P}(z, \kappa, \psi_{inc}, \omega)] [\mathbf{f}(z, \kappa, \psi_{inc}, \omega)]. \quad (9)$$

In this equation,

$$[\mathbf{f}(z, \kappa, \psi_{inc}, \omega)] = [e_x(z, \kappa, \psi_{inc}, \omega), e_y(z, \kappa, \psi_{inc}, \omega), h_x(z, \kappa, \psi_{inc}, \omega), h_y(z, \kappa, \psi_{inc}, \omega)]^T \quad (10)$$

is a column 4-vector, and $[\mathbf{P}(z, \kappa, \psi_{inc}, \omega)]$ is a 4×4 matrix function of z that can be easily obtained using symbolic manipulation programs such as Mathematica.

Analytic solution of (9) can be obtained, provided $[\mathbf{P}(z, \kappa, \psi_{inc}, \omega)] = [\mathbf{P}_{con}(\kappa, \psi_{inc}, \omega)]$ is not a function of z . This happens, of course, for columnar thin films [11], and the solution procedure is described by Hochstadt [12]. Exact analytic solution of (9) has been obtained also for axial propagation (i.e., $\kappa = 0$) in periodic TFHBMs [14]; and a solution in terms of a convergent matrix polynomial series is available for oblique propagation (i.e., $\kappa \neq 0$) in periodic TFHBMs [15].

More generally, only a numerical solution of (9) can be obtained. Suppose that $[\mathbf{P}(z, \kappa, \psi_{inc}, \omega)]$ is a periodic function of z . Then, a perturbative approach can be used to obtain simple results for weakly periodic STFs [16, 17], while a coupled-mode approach may come handy if otherwise [18]. But if $[\mathbf{P}(z, \kappa, \psi_{inc}, \omega)]$ is not periodic, the constitutive dyadics can assumed to be piecewise homogeneous over slices of thickness Δz , and the approximate transfer equation [12]

$$[\mathbf{f}(z + \Delta z, \kappa, \psi_{inc}, \omega)] \simeq \exp \left\{ i[\mathbf{P}(z + \frac{\Delta z}{2}, \kappa, \psi_{inc}, \omega)] \Delta z \right\} [\mathbf{f}(z, \kappa, \psi_{inc}, \omega)] \quad (11)$$

can be suitably manipulated with appropriately small values of Δz .

3. From the nanoscopic to the continuum

3.1 Nominal model

Equations (1) and (2) incorporate the assumption of a STF as a unidirectionally nonhomogeneous continuum. This is valid in a macroscopic sense, i.e., when the length scale of the film morphology is considerably smaller than the electromagnetic probe wavelength. This assumption holds true in the visible and the infrared frequency regimes, because STFs with appropriate morphological length scales can be fabricated.

Let us consider a simple nanoscopic-to-macroscopic homogenization formalism to determine the reference constitutive dyadics $\underline{\underline{\epsilon}}_{ref}(\omega)$, etc. [19]. The chosen STF is made of a bianisotropic material, whose bulk constitutive relations are specified as

$$\left. \begin{aligned} \mathbf{D}(\mathbf{r}, \omega) &= \epsilon_0 [\underline{\underline{\epsilon}}_s(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\alpha}}_s(\omega) \cdot \mathbf{H}(\mathbf{r}, \omega)] \\ \mathbf{B}(\mathbf{r}, \omega) &= \mu_0 [\underline{\underline{\beta}}_s(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\mu}}_s(\omega) \cdot \mathbf{H}(\mathbf{r}, \omega)] \end{aligned} \right\}. \quad (12)$$

The void regions of the STF are taken to be occupied by a medium with the following bulk properties:

$$\left. \begin{aligned} \mathbf{D}(\mathbf{r}, \omega) &= \epsilon_0 [\underline{\underline{\epsilon}}_v(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\alpha}}_v(\omega) \cdot \mathbf{H}(\mathbf{r}, \omega)] \\ \mathbf{B}(\mathbf{r}, \omega) &= \mu_0 [\underline{\underline{\beta}}_v(\omega) \cdot \mathbf{E}(\mathbf{r}, \omega) + \underline{\underline{\mu}}_v(\omega) \cdot \mathbf{H}(\mathbf{r}, \omega)] \end{aligned} \right\}. \quad (13)$$

Both mediums are supposed to be present in the STF as ellipsoids which are entirely notional. In every xy plane, the longest axes of all ellipsoids of both mediums should be aligned in parallel. The shapes of the two types of ellipsoids can be different, the respective surfaces being defined by the functions

$$\mathbf{r}_s(\theta, \phi) = \delta_s \underline{\underline{U}}_s \cdot \mathbf{u}_r(\theta, \phi), \quad \mathbf{r}_v(\theta, \phi) = \delta_v \underline{\underline{U}}_v \cdot \mathbf{u}_r(\theta, \phi). \quad (14)$$

Here, $\mathbf{u}_r(\theta, \phi)$ is the radial unit vector in a spherical coordinate system located at the centroid of an ellipsoid; the scalars $\delta_{s,v}$ are linear measures of the ellipsoidal sizes; and the shape dyadics $\underline{\underline{U}}_{s,v}$ are real dyadics of rank 3, with positive eigenvalues $0 < a_{s,v}^{(j)} \leq 1$, ($j = 1, 2, 3$). Setting $a^{(3)} \gg a^{(1)}$ and $a^{(3)} \gg a^{(2)}$ will make a particular ellipsoid almost like a needle with a slight bulge in its middle part. The porosity of the STF is denoted by f_v , ($0 \leq f_v \leq 1$).

The use of 6×6 dyadics provides notational simplicity for treating electromagnetic fields in bianisotropic materials. Thus, we define the 6×6 dyadics

$$\underline{\underline{\mathbf{C}}}_{ref,s,v} = \left(\begin{array}{c|c} \epsilon_0 \underline{\underline{\epsilon}}_{ref,s,v} & \epsilon_0 \underline{\underline{\alpha}}_{ref,s,v} \\ \hline \mu_0 \underline{\underline{\beta}}_{ref,s,v} & \mu_0 \underline{\underline{\mu}}_{ref,s,v} \end{array} \right). \quad (15)$$

The ω -dependences of various quantities are not explicitly mentioned in this and the following equations for compactness.

The celebrated Bruggeman formalism is now implemented to effect local homogenization (with reference to any xy plane) [19]. For this purpose, the 6×6 polarizability dyadics

$$\underline{\underline{\mathbf{a}}}_{s,v} = \left(\underline{\underline{\mathbf{C}}}_{s,v} - \underline{\underline{\mathbf{C}}}_{ref} \right) \cdot \left[\underline{\underline{\mathbf{I}}} + i\omega \underline{\underline{\mathbf{D}}}_{s,v} \cdot \left(\underline{\underline{\mathbf{C}}}_{s,v} - \underline{\underline{\mathbf{C}}}_{ref} \right) \right]^{-1} \quad (16)$$

are defined, where $\underline{\underline{\mathbf{I}}}$ is the 6×6 identity dyadic. The 6×6 depolarization dyadics $\underline{\underline{\mathbf{D}}}_{s,v}$ must be

computed *via* two-dimensional integration as follows:

$$\underline{\underline{\mathbf{D}}}_{s,v} = \frac{1}{4\pi i \omega \epsilon_0 \mu_0} \int_{\phi=0}^{2\pi} d\phi \int_{\theta=0}^{\pi} d\theta \sin \theta \frac{(\underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r)(\mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger})}{A_{s,v}(\mathbf{u}_r)} \times \left(\begin{array}{c|c} \mu_0 \mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\mu}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r & -\epsilon_0 \mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\alpha}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r \\ \hline -\mu_0 \mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\beta}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r & \epsilon_0 \mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\epsilon}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r \end{array} \right). \quad (17)$$

Here, $\mathbf{u}_r = \mathbf{u}_r(\theta, \phi)$ is the unit radial vector,

$$A_{s,v}(\mathbf{u}_r) = (\mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\epsilon}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r)(\mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\mu}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r) - (\mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\alpha}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r)(\mathbf{u}_r \cdot \underline{\underline{U}}_{s,v}^{\dagger} \cdot \underline{\underline{\beta}}_{ref} \cdot \underline{\underline{U}}_{s,v}^{-1} \cdot \mathbf{u}_r) \quad (18)$$

while $\underline{\underline{U}}_s^{\dagger}$ is the transpose of $\underline{\underline{U}}_s^{-1}$, etc. The Bruggeman formalism then consists of the solution of the equation [20, 21]

$$f_v \underline{\underline{\mathbf{a}}}_v + (1 - f_v) \underline{\underline{\mathbf{a}}}_s = \underline{\underline{\mathbf{0}}}, \quad (19)$$

with $\underline{\underline{\mathbf{0}}}$ as the 6×6 null dyadic. This equation has to be numerically solved for $\underline{\underline{\mathbf{C}}}_{ref}$, and a Jacobi iteration technique is recommended for that purpose [21, 22].

The solution of (19) represents the homogenization of objects of microscopic linear dimensions to a macroscopic continuum. The quantities entering $\underline{\underline{S}}(z)$ are known prior to fabrication, as also are χ , $\underline{\underline{\mathbf{C}}}_s$ and $\underline{\underline{\mathbf{C}}}_v$. In order to calibrate the nominal model presented, the shape dyadics $\underline{\underline{U}}_s$ and $\underline{\underline{U}}_v$ can be chosen by comparison of the predicted $\underline{\underline{\mathbf{C}}}_{ref}$ against measured data.

3.2 Application to dielectric TFHBMs

Sherwin and Lakhtakia [22] used the foregoing model to extensively study the planewave responses of dielectric TFHBMs, with $\underline{\underline{\mu}}_{s,v} = \underline{\underline{I}}$, $\underline{\underline{\alpha}}_{s,v} = \underline{\underline{0}}$, $\underline{\underline{\beta}}_{s,v} = \underline{\underline{0}}$, $\underline{\underline{\epsilon}}_s = \epsilon_s \underline{\underline{I}}$, and $\underline{\underline{\epsilon}}_v = \underline{\underline{I}}$. With λ_0 denoting the wavelength in vacuum, a Lorentz resonance model was chosen for ϵ_s as per

$$\epsilon_s = 1 + \frac{q_s}{1 + (N_s^{-1} - i\lambda_s \lambda_0^{-1})^2}, \quad (20)$$

with constants q_s , N_s and λ_s selected so that absorption is moderate for visible wavelengths. The ellipsoids were chosen to be identical (i.e., $\underline{\underline{U}}_s = \underline{\underline{U}}_v$), as described by

$$(\mathbf{r} \cdot \mathbf{u}_n)^2 + \left(\frac{\mathbf{r} \cdot \mathbf{u}_b}{\gamma_2} \right)^2 + \left(\frac{\mathbf{r} \cdot \mathbf{u}_\tau}{\gamma_3} \right)^2 = \delta^2, \quad (21)$$

where the transverse aspect ratio $\gamma_2 > 1$ and the slenderness ratio $\gamma_3 \gg 1$ relate the three principal axes.

For normally incident planewaves and with

$$\underline{\underline{S}}(z) = \underline{\underline{S}}_z(z) \Big|_{\zeta(z)=\pi z/\Omega} = \mathbf{u}_z \mathbf{u}_z + (\mathbf{u}_x \mathbf{u}_x + \mathbf{u}_y \mathbf{u}_y) \cos \frac{\pi z}{\Omega} + (\mathbf{u}_y \mathbf{u}_x - \mathbf{u}_x \mathbf{u}_y) \sin \frac{\pi z}{\Omega}, \quad (22)$$

the spectrums of optical rotation, transmittance ellipticity, linear dichroism, circular dichroism, apparent linear dichroism, and apparent circular dichroism were calculated as functions of the constitutive and the geometric parameters ϵ_s , γ_2 , γ_3 , χ , Ω , $f = 1 - f_v$, and λ_0 . Maximum magnitudes in the computed spectrums were determined in specific wavelength-regimes. The variations of these maximums were then examined with respect to any one of the constitutive and the geometric parameters, while the other parameters were held fixed. From these studies, the following significant conclusions were reached:

- (i) All observable response properties strongly depend on the transverse aspect ratio γ_2 , $1 \leq \gamma_2 \ll \gamma_3$. There exists a specific value of γ_2 denoted by γ_2^o such that all optical activity disappears. The value of γ_2^o can be parameterized in terms of other geometric factors and ϵ_s .
- (ii) All observable property maximums are best-fitted to fourth-order polynomials of γ_2 .
- (iii) All observable property maximums have similar dependencies on the volume fraction f ; furthermore, $p^{max} \rightarrow 0$ as $f \rightarrow 0, 1$. (Here, p^{max} denotes the maximum magnitude of the observable response property p over a prescribed range of λ_0 .)
- (iv) There exists an f_o for each value of γ_2 such that $p^{max}(f_o) > p^{max}(f)$, $f \neq f_o$. The value of f_o depends on the values of other geometric and constitutive parameters as well as on the property p . An increase in γ_2 results in a decrease in f_o .

Details of these and other results will appear in print shortly [23], and the identified functional relationships should assist in design of STF-based devices as well as the on-line monitoring of STF fabrication processes.

4. Optical applications

Although many applications are possible [5], the potential of STFs has been most successfully exploited for optical filters. Chiral STFs, which are appropriately described as the periodic dielectric TFHBMs of Section 3.2, must display the circular Bragg phenomenon in accordance with their periodic nonhomogeneity along the z axis [6, 24], thereby ensconcing themselves firmly in the area of *chiral optics*. Briefly, a structurally right- (resp. left-) handed chiral STF only a few periods thick almost completely reflects axially incident, right (resp. left) circularly polarized light with wavelength lying in the so-called Bragg regime; while the reflection of axially incident, left (resp. right) circularly polarized light in the same regime is very little. The bandwidth of the Bragg regime and the peak reflectivity therein first increase with the thickness of the chiral STF, and then saturate. Once this saturation has occurred, further thickening of the film has negligible effects on the reflection spectrum.

4.1 Circular polarization filters

The circular Bragg phenomenon can be employed to realize circular polarization filters. Normally incident, circularly polarized light of one handedness can be reflected almost completely, while that of the other handedness is substantially transmitted, if absorption is small enough and the film is sufficiently thick, in the Bragg regime. This was demonstrated by Wu *et al.* [25] with chiral STFs fabricated using the serial bideposition technique. As of now, the Bragg regime can be positioned at virtually any $\lambda_0 \in [450, 1700]$ nm. Polarization insensitivity can be realized using a bilayer version, as a cascade of two otherwise identical chiral STFs but of opposite structural handedness [26]. Chirping can be used to widen the bandwidth [27], and tightly interlaced chiral STFs may also hold technological attraction [28].

4.2 Polarization–discriminatory handedness–inverters

A polarization–discriminatory handedness–inverter for circularly polarized light was fabricated using STF technology. This was the first realization of a two–section STF device. It comprises a chiral STF [24] and a half–wave plate realized as a columnar thin film [11]. Basically, it almost completely reflects, say, left circularly polarized light; while it substantially transmits incident right circularly polarized light after transforming it into left circularly polarized light. Theoretical predictions [29] were borne out experimentally [30].

4.3 Spectral hole filters

In a further bid to illustrate the potential of the STF concept, a three–section STF was proposed as a spectral hole filter. Its first and third layers are identical chiral STFs, while the thin middle layer is homogeneous [31, 32]. The middle layer is supposed to act as a phase defect. This design was actually implemented to obtain a 11 nm wide spectral hole centered at $\lambda_0 = 580$ nm [33]. The realized bandwidth filter compares very favorably with those of commercially available holographic filters.

A better design became available shortly thereafter and was experimentally evaluated too [34]. The middle layer was eliminated, but the lower chiral STF was twisted by 90° with respect to the upper chiral STF about the z axis. The twist acts as the required phase defect.

4.4 Rugate and Šolc filters

SNTFs can also be pressed into service as optical filters — for linearly polarized plane waves. McPhun *et al.* [35] fabricated rugate filters with STF technology for narrow–band reflection applications. Šolc filters of the *fan* and the *folded* types are also possible with the same technology [36].

4.5 Fluid sensors

The porosity of STFs makes them attractive for fluid concentration sensing applications [37, 38], because their optical response properties must change in accordance with the number density of molecules intruding into the void regions. In particular, theoretical research has shown that the Bragg regime of a chiral STF must shift accordingly, thereby providing a measure of the

fluid concentration [37]. Qualitative support for this finding is provided by experiments on wet and dry chiral STFs [39].

Very recent theoretical research has indicated that STF spectral hole filters can function as highly sensitive fluid concentration sensors; and proof-of-concept experiments with both circularly polarized and unpolarized incident light have confirmed the red-shift of spectral holes upon exposure to moisture [40].

5. Emerging applications

From their inception [5], STFs were expected to have a wide range of applications, implementable only after their optical, electronic, and magnetic properties came to better understood. Their optical applications came to be investigated first, as detailed in Section 4. However, their high porosity — in combination with optical anisotropy and possible two-dimensional electron confinement in the microstructure — makes STFs potential candidates as

- (i) electroluminescent devices (emitting light of a pre-specified polarization state from circular to linear) prepared by chemical vapor deposition of nanocrystal silicon into the void spaces of STFs formed from wide-gap transparent oxides;
- (ii) high speed, high efficiency electrochromic films;
- (iii) optically transparent conducting films sculptured from pure metals;
- (iv) multi-state electronic switches based on filamentary conduction;
- (v) optical sensors that can detect and quantify various chemical and biological fluids; and
- (vi) micro-sieves for the entrapment of viruses or for growing biological tissues on surfaces of biological or non-biological provenances.

Obviously, many other applications may turn out to be possible, but significant progress thus far has been reported, to my knowledge, only in the following five areas:

5.1 Biochips

Endowed with porosity of engineered texture, STFs can function as microreactors and therefore can function as biochips. As an example, let us consider the following scenario: Intercalation of a ruthenium complex with double-stranded DNA is known to generate luminescence. Suppose that identical single-stranded DNA molecules — with a particular genomic sequence matched to, say, *E. coli* — are dispersed in a STF. A drop of contaminated water containing analyte DNA molecules from exploded *E. coli* is put on the STF, followed by a drop of an appropriate ruthenium complex. The bioluminescence signal emerging from the STF can be optically sensed by photon counters in order to measure the degree of contamination.

Bioluminescent emission is bound to be affected by the reactor characteristics. If the reactor is a chiral STF, the possibility of exploiting the circular Bragg phenomenon exhibited by it

would be attractive. Indeed, the structural handedness as well as the periodicity of chiral STFs have been shown to critically control the emission spectrum and intensity, while the polarization state of the emitted light is strongly correlated with the structural handedness of the embedded source filaments [41]. Bioluminescence STF sensors therefore merit closer attention.

5.2 *Optical interconnects*

Efficient use of optoelectronic devices requires the development of optical interconnects which, in addition to providing effective signal transmission, must be simple to fabricate on integrated circuitry. STF technology is compatible with the planar technology of electronic chips. Guided wave propagation in chiral STFs turns out to yield the space-guide concept: the capacity to simultaneously support propagation modes with different phase velocities in different directions [42, 43]. This could result in efficient use of the available *real estate* in electronic chips. Furthermore, the helicoidal microstructure of chiral STFs would resist vertical cleavage and fracture, in contrast to columnar thin films which can also function as space-guides.

Chiral STFs can be grown as a regular lattice by lithographically patterning the substrates [10]. Whereas slow substrate-rotation rates result in the growth of arrays of microhelixes or microsprings spaced as close as 20 nm from their nearest neighbors, faster rotation rates yield arrays of increasingly denser pillars [9, 10]. Such STFs are essentially photonic bandgap materials in the visible and the infrared regimes. Most recently, even line defects have been introduced therein [44].

5.3 *Interlayer dielectrics*

With the microelectronics industry moving relentlessly towards decreasing feature sizes and increasingly stringent tolerance levels, an urgent need exists for the use of low-permittivity materials as interlevel dielectrics. Silicon dioxide, the current material of choice, has excellent properties in all respects except one: its permittivity is too high. The porosity of STFs and nanoporous silica makes them attractive low-permittivity materials for microelectronic and electronic packaging applications. However, chiral STFs are likely to have significant thermal, mechanical, as well as electrical advantages over nanoporous silica — because of (i) porosity with controllable texture and (ii) helicoidal morphology. Also, chiral STFs could be impregnated with various kinds of polymers [45].

5.4 *Electrically addressable displays*

Liquid crystals (LCs) can be electronically addressed [46] and are therefore widely used these days for displays. Although STFs are not electronically addressable, the alignment of nematic LCs forced into the void regions of chiral STFs has been shown to respond to applied voltages [47]. Thus, STF-LC composites may have a future as robust displays.

5.5 *Optical pulse-shapers*

The current explosive growth of digital optics communication has provided impetus for time-

domain research on novel materials. As chiral STFs are very attractive for optical applications, the circular Bragg phenomenon is being studied in the time domain. A pulse bleeding phenomenon has been identified as the underlying mechanism, which can drastically affect the shapes, amplitudes and spectral components of femtosecond pulses [48]. However, narrow-band rectangular pulses can pass through without significant loss of information [49]. Application of STFs to shape optical pulses appears to be waiting in the wings.

6. Concluding remarks

The foregoing section makes it evident that sculptured thin films are comparable to liquid crystals [45, 50, 51] in many respects. Their respective roles can be competitive as well as complementary, depending on the specific environment and application. For instance, being soft and viscous, LCs serve as pressure/temperature sensors. But STFs are porous solids and likely to be unaffected by small changes in the ambient pressure and temperature. However, those qualities are likely to be useful in certain environments where mechanical integrity and thermal stability are at a premium. Furthermore, STFs can serve as fluid sensors and microreactors, but LCs can not. Liquid crystals are widely used for electronic displays as they are electrically addressable; STF-LC composites can also be electrically addressed. Both LCs and STFs can be used as optical filters and polarizers, but only the latter truly make the concept of optics-in-a-chip possible. Finally, the wide scope of STFs — accessible through tailorable microstructure and anisotropy, as well as *via* the almost unlimited types of depositable materials — is remarkable.

To conclude, the development of STF technology is now in a post-embryonic stage. Much needs to be done to make it robust, economical and widely used. But the future appears bright, and the recent feat of Suzuki & Taga [52] in fabricating STFs with many sections underscores the tremendous promise of STFs as integrated optical chips [5, 12].

Acknowledgements. I am indebted to all of my collaborators and my students for splendid support on STF research over the last decade. This review is dedicated to the inspirational batting performances of V.V.S. Laxman and Rahul S. Dravid and the superb bowling performance of Harbhajan Singh during a cricket Test match between India and Australia, played March 11–15, 2001 at Eden Gardens, Kolkata.

7. References

- [1] A. Lakhtakia, W.S. Weiglhofer, *Microw. Opt. Technol. Lett.* 6 (1993) 804.
- [2] A. Lakhtakia, R. Messier, in: F. Mariotte, J.-P. Parneix (Eds.), *Proceedings of Chiral '94* (Périgueux, France, May 18–20, 1994), French Atomic Energy Commission, Le Barp, France, 1994, pp. 125–130.

- [3] K. Robbie, M.J. Brett, A. Lakhtakia, *J. Vac. Sci. Technol. A* 13 (1995) 2991.
- [4] N. O. Young, J. Kowal, *Nature* 183 (1959) 104.
- [5] A. Lakhtakia, R. Messier, M.J. Brett, K. Robbie, *Innov. Mater. Res.* 1 (1996) 165.
- [6] V.C. Venugopal, A. Lakhtakia, in: O.N. Singh, A. Lakhtakia (Eds.), *Electromagnetic Fields in Unconventional Materials and Structures*, Wiley, New York, 2000, pp. 151–216.
- [7] A. Lakhtakia, R.F. Messier (Eds.), *Engineered Nanostructural Films and Materials*, SPIE, Bellingham, WA, USA, 1999.
- [8] I. Hodgkinson, Q.H. Wu, *Appl. Opt.* 38 (1999) 3621.
- [9] R. Messier, V.C. Venugopal, P.D. Sunal, *J. Vac. Sci. Technol. A* 18 (2000) 1538.
- [10] M. Malac, R.F. Egerton, *Nanotechnology* 12 (2001) 11.
- [11] I.J. Hodgkinson, Q.-h. Wu, *Birefringent Thin Films and Polarizing Elements*, World Scientific, Singapore, 1997.
- [12] A. Lakhtakia, *Optik* 107 (1997) 57.
- [13] H. Hochstadt, *Differential Equations — A Modern Approach*, Dover Press, New York, 1975, chap. 2.
- [14] A. Lakhtakia, W.S. Weiglhofer, *Proc. R. Soc. Lond. A* 448 (1995) 419; erratums: 454 (1998) 3275.
- [15] A. Lakhtakia, W.S. Weiglhofer, *Proc. R. Soc. Lond. A* 453 (1997) 93; erratums: 454 (1998) 3275.
- [16] V.A. Yakubovich, V.M. Starzhinskii, *Linear Differential Equations with Periodic Coefficients*, Wiley, New York, 1975.
- [17] W.S. Weiglhofer, A. Lakhtakia, *Optik* 102 (1996) 111.
- [18] K. Rokushima, J. Yamakita, *J. Opt. Soc. Am. A* 4 (1987) 27.
- [19] A. Lakhtakia, P.D. Sunal, V.C. Venugopal, E. Ertekin, *Proc. SPIE* 3790 (1999) 77.
- [20] W.S. Weiglhofer, A. Lakhtakia, B. Michel, *Microw. Opt. Technol. Lett.* 15 (1997) 263; erratum: 22 (1999) 221.
- [21] B. Michel, in: O.N. Singh, A. Lakhtakia (Eds.), *Electromagnetic Fields in Unconventional*

Materials and Structures, Wiley, New York, 2000, pp. 39–82.

- [22] J.A. Sherwin, A. Lakhtakia, Proc. SPIE 4097 (2000) 250.
- [23] J.A. Sherwin, A. Lakhtakia, Math. Comput. Model. (accepted for publication in 2001).
- [24] I. Hodgkinson, Q.h. Wu, B. Knight, A. Lakhtakia, K. Robbie, Appl. Opt. 39 (2000) 642.
- [25] Q. Wu, I.J. Hodgkinson, A. Lakhtakia, Opt. Eng. 39 (2000) 1863.
- [26] A. Lakhtakia, V.C. Venugopal, Microw. Opt. Technol. Lett. 17 (1998) 135.
- [27] A. Lakhtakia, Microw. Opt. Technol. Lett. 28 (2001) 323.
- [28] A. Lakhtakia, Optik 112 (2001) 119.
- [29] A. Lakhtakia, Opt. Eng. 38 (1999) 1596.
- [30] I.J. Hodgkinson, A. Lakhtakia, Q.h. Wu, Opt. Eng. 39 (2000) 2831.
- [31] A. Lakhtakia, M. McCall, Opt. Commun. 168 (1999) 457.
- [32] A. Lakhtakia, V.C. Venugopal, M.W. McCall, Opt. Commun. 177 (1999) 57.
- [33] I.J. Hodgkinson, Q.h. Wu, A. Lakhtakia, M.W. McCall, Opt. Commun. 177 (2000) 79.
- [34] I.J. Hodgkinson, Q.H. Wu, K.E. Thorn, A. Lakhtakia, M.W. McCall, Opt. Commun. 184 (2000) 57.
- [35] A.H. McPhun, Q.H. Wu, I.J. Hodgkinson, Electron. Lett. 34 (1998) 360.
- [36] A. Lakhtakia, Opt. Eng. 37 (1998) 1870.
- [37] A. Lakhtakia, Sensors & Actuators B: Chem. 52 (1998) 243.
- [38] E. Ertekin, A. Lakhtakia, Eur. Phys. J. Appl. Phys. 5 (1999) 45.
- [39] I.J. Hodgkinson, Q.h. Wu, K.M. McGrath, Proc. SPIE 3790 (1999) 184.
- [40] A. Lakhtakia, M.W. McCall, J.A. Sherwin, Q.h. Wu, I.J. Hodgkinson, Opt. Commun. (accepted for publication in 2001).
- [41] A. Lakhtakia, Opt. Commun. 188 (2001) 313.
- [42] A. Lakhtakia, Optik 110 (1999) 289.

- [43] E. Ertekin, A. Lakhtakia, Proc. R. Soc. Lond. A 457 (2001) 817.
- [44] M. Malac, R.F. Egerton, J. Vac. Sci. Technol. A 19 (2000) 158.
- [45] V.C. Venugopal, A. Lakhtakia, R. Messier, J.-P. Kucera, J. Vac. Sci. Technol. B 18 (2000) 32.
- [46] S.D. Jacobs (Ed.), Selected Papers on Liquid Crystals for Optics, SPIE, Bellingham, WA, USA, 1992.
- [47] J.C. Sit, D.J. Broer, M.J. Brett, Liq. Cryst. 27 (2000) 387.
- [48] J.B. Geddes III, A. Lakhtakia, Eur. Phys. J. Appl. Phys. 13 (2001) 3.
- [49] J.B. Geddes III, A. Lakhtakia, Microw. Opt. Technol. Lett. 28 (2001) 59.
- [50] S. Chandrasekhar, Liquid Crystals, Cambridge University Press, Cambridge, UK, 1992.
- [51] P.G. de Gennes, J. Prost, The Physics of Liquid Crystals, Clarendon Press, Oxford, UK, 1993.
- [52] M. Suzuki, Y. Taga, Jap. J. Appl. Phys. 40, part 2 (2001) L358.